Magnetic field induced rotation of the d-vector in the spin triplet superconductor Sr_2RuO_4

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In zero magnetic field the superconductor Sr_2RuO_4 is believed to have a chiral spin triplet pairing state in which the gap function d-vector is aligned along the crystal c-axis. Using a phenomenological but orbital specific description of the spin dependent electron-electron attraction and a realistic quantitative account of the electronic structure in the normal state we analyze the orientation of the spin triplet Cooper pair d-vector in response to an external c-axis magnetic field. We show that for suitable values of the model parameters a c-axis field of only 20 mT is able to cause a reorientation phase transition of the d-vector from along c to the $a-b$ plane, in agreement with recent experiments.

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INTRODUCTION

The 1.5 K superconductor Sr_2RuO_4 [1] is widely believed to be a rare example of a spin-triplet Cooper paired system [2, 3]. Unlike heavy fermion materials which are possible candidates for spin triplet pairing (such as UPt_3) there are no $4f$ or $5f$ heavy elements in the unit cell, and so the effects of spin-orbit coupling should be relatively weak. In this respect paring in Sr_2RuO_4 should be closely analogous to the case of superfluid ^{3}He. One should expect that weak or moderate B-fields would be able to rotate the d-vector order parameter and induce domain walls, textures and other topological defects. As in the case of ^3He, the experimental observation of such d-vector rotations would be a decisive test of the pairing symmetry. Systematic study of such rotations as a function of various physical parameters would both confirm the pairing symmetry and also place strong constraints on the pairing mechanism and microscopic Hamiltonian parameters.

One of the strongest pieces of evidence for spin triplet Cooper pairing in Sr_2RuO_4 was the observation that the electronic spin susceptibility $\chi_s(T)$ remained constant below T_c for magnetic fields in the a-b plane [4, 5]. This observation is inconsistent with a spin-singlet pairing state (s or d-wave) for which $\chi_s(T) = \chi_n Y(T)$, where χ_n is the normal state Pauli spin susceptibility and $Y(T)$ is the Yoshida function. On the other hand the observations would be immediately consistent with a chiral symmetry paring state of the form

$$\mathbf{d}(\mathbf{k}) = (\sin k_x + i \sin k_y)\hat{\mathbf{e}}_z, \quad (1)$$

which below we shall refer to as pairing state (a). For such a pairing state is is well known [6] that the spin susceptibility has the following tensor form

$$\hat{\chi}_s(T) = \chi_n \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & Y(T) \end{pmatrix}. \quad (2)$$

and so the susceptibility tensor is constant as a function of T for field directions perpendicular to the $\mathbf{d}$ vector. The ^{17}O Knight shift experiments performed in (ab) plane magnetic fields [4] and neutron scattering experiments [5] both observed a constant susceptibility below T_c, a result which *uniquely* points to a chiral triplet paring state with $\mathbf{d} \parallel \hat{\mathbf{e}}_z$.

More recently Murakawa *et al.* [7] have measured the ^{101}Ru-Knight shift of Sr_2RuO_4 in a superconducting state under the influence of magnetic field parallel to the c-axis. In contradiction to expectations from Eqs. (1) and (2) they found that its value is *also unchanged from the normal state value below T_c*. They remarked that this result would be consistent with Eq. (2) only if it is assumed that the $\mathbf{d}$ vector rotates away from the $\hat{\mathbf{e}}_\mathbf{z}$ direction under the influence of the external field. Such a rotation of the $\mathbf{d}$ vector is well known in the case of superfluid ^{3}He-A [6], where to minimize free energy $\mathbf{d}$ orients itself perpendicular to the external field unless pinned by surface effects. However, in Sr_2RuO_4 it is expected that spin-orbit coupling would fix the chiral state $\mathbf{d}$ vector to the crystal c-axis [8, 9, 10, 11]. Murakawa *et al.* [7] were able to measure $\chi_s(T)$ down to fields as low as 20 mT at 80 mK, showing that it was equal to χ_n to within experimental accuracy at all temperatures and fields measured below B_{c2} (75 mT for c-axis fields). The implication of this result, that $\mathbf{d}$ vector rotation must occur at below 20 mT, therefore places very strong constraints on the strength of pinning by the spin-orbit coupling.

In this work we examine the combined effects of spin-orbit coupling and c-axis magnetic field on the chiral

state of Sr_2RuO_4. We show that a realistic physical model, which we have previously shown to be consistent with a wide range of experimental data, allows us to find reasonable parameter ranges where the chiral state Eq. (1) is stable in zero field, but where a transition to another state occurs even for fields of order 20 mT or smaller. The minimum energy states in finite c-axis field are not simply the chiral state with $\mathbf{d}$ vector rotated to the $a-b$ plane, but rather are non-chiral pairing states of the form

$$\begin{aligned} \mathbf{d}(\mathbf{k}) &= (\sin k_x, \sin k_y, 0) \\ \mathbf{d}(\mathbf{k}) &= (\sin k_y, -\sin k_x, 0) \end{aligned} \quad (3)$$

on the γ Fermi surface sheet (which below we refer to as (b) and (c) respectively). The spin susceptibility for either of these states is of the form [6, 12]

$$\hat{\chi}_s(T) = \frac{1}{2}\chi_n \begin{pmatrix} 1+Y(T) & 0 & 0 \\ 0 & 1+Y(T) & 0 \\ 0 & 0 & 2 \end{pmatrix}, \quad (4)$$

corresponding to constant spin susceptibility for c-axis fields consistent with the results of Murakawa *et al.* [7].

Since these states are symmetry distinct from the chiral state Eq. (1) the transition from one to another should occur at a finite external field, B_t, and it should be accompanied by a finite entropy change ΔS. Below we map out a generic phase diagram for the transitions between pairing states of Sr_2RuO_4 in a c-axis field, assuming parameter values consistent with the Murakawa's *et al.* experiments [7]. We also estimate the entropy change associated with the transition, and comment on its experimental observability. Our preliminary results have been presented in [13].

THE MODEL

As the pairing mechanism operating in the strontium ruthenate superconductor is not known [2, 3] it is important to understand the experiments on the basis of semi-phenomenological models [14, 15]. Specifically we write the effective pairing Hamiltonian as

$$\begin{aligned} \hat{H} &= \sum_{ijmm',\sigma} \left((\varepsilon_m - \mu)\delta_{ij}\delta_{mm'} - t_{mm'}(ij) \right) \hat{c}^{+}_{im\sigma}\hat{c}_{jm'\sigma} \\ &\quad + \mathrm{i}\frac{\lambda}{2} \sum_{i,\sigma\sigma'\kappa} \sum_{mm'} \varepsilon^{\kappa m m'} \sigma^{\kappa}_{\sigma\sigma'} c^{+}_{im\sigma} c_{im'\sigma'} \\ &\quad - \frac{1}{2} \sum_{ijmm'} \sum_{\alpha\beta\gamma\delta} U^{\alpha\beta,\gamma\delta}_{mm'}(ij) \hat{c}^{+}_{im\alpha}\hat{c}^{+}_{jm'\beta}\hat{c}_{jm'\gamma}\hat{c}_{im\delta}, \end{aligned} \quad (5)$$

where m and m' refer to the three Ru t_{2g} orbitals $a = d_{xz}$, $b = d_{yz}$ and $c = d_{xy}$, and i and j label the sites of a body centered tetragonal lattice. The hopping integrals $t_{mm'}(ij)$ and site energies ε_m were fitted to reproduce the experimentally determined Fermi surface [16, 17]. λ is the effective Ru 4d spin-orbit coupling parameter, and the effective Hubbard parameters $U^{\alpha\beta,\gamma\delta}_{mm'}(ij)$ are generally both spin [18, 19] and orbital dependent [14, 15, 20].

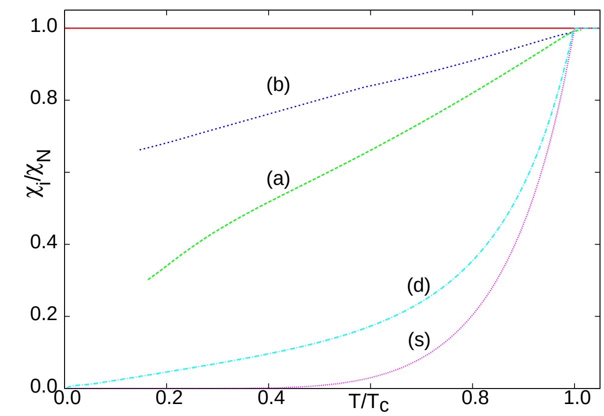

FIG. 1: (Color online) Temperature dependence of the normalized spin susceptibility in the superconducting state for a number of order parameters. The curves marked (a) and (b) show the c-axis susceptibility for Sr_2RuO_4 and chiral symmetry state, (a), defined by Eq. 6 and Eq. 7, and the ab-plane susceptibility of the non-chiral triplet state, of symmetry (b), corresponding to Eq. 3. Curves labelled (s) and (d) show the Yoshida function for single band superconductor with two dimensional tight binding spectrum and order parameter of s and d wave symmetry, respectively.

Our approach is based on self consistent solution of the Bogolubov-deGennes equations for the model (5) in the spin triplet channel for each possible symmetry distinct order parameter. We have shown elsewhere [20, 21] that a minimal realistic model requires two Hubbard U parameters: $U_{\parallel}$ for nearest neighbor in-plane interactions between Ru d_{xy} orbitals (corresponding to the γ Fermi surface sheet) and $U_{\perp}$ for out of plane nearest neighbor interactions between Ru d_{xz} and d_{yz} orbitals (corresponding to the α and β Fermi surface sheets). When these two parameters are chosen to be spin independent constants (for $\lambda = 0$) and to give the experimental $T_c = 1.5$K on all three α, β and γ Fermi surface sheets, then the calculated specific heat, thermal conductivity and $a-b$ plane penetration depth are in very good agreement with experiments [22, 23, 24].

Figure (1) shows the susceptibilities calculated within the model for the (a) and (b) states with the symmetries given by Eqs. 1 and 3. For the chiral state (a) the triplet d-vector is defined in all 3 bands as $\mathbf{d}(\mathbf{k}) = \Delta_{mm'}(\mathbf{k})\hat{\mathbf{e}}_z$, with $\Delta_{mm'}(\mathbf{k})$ denoting contributions from different orbitals which are given by

$$\Delta_{cc}(\mathbf{k}) = \Delta_{cc}(T)(\sin k_x + i \sin k_y) \quad (6)$$

for the Ru $c(= d_{xy})$ orbitals and,

$$\Delta_{mm'}(\mathbf{k}) = \Delta_{mm'}(\sin\frac{k_x}{2}\cos\frac{k_y}{2} + i\sin\frac{k_y}{2}\cos\frac{k_y}{2})\cos\frac{k_z c}{2} \quad (7)$$

for $m, m' = a, b$ corresponding to the Ru d_{xz} and d_{yz} orbitals [20]. The susceptibility results, for the chiral (a) state show that the dependence of χ for c-axis B-fields is is relatively structureless. The relevant Yoshida function $Y(T)$ is equal to unity at $T = T_c$ and drops essentially linearly to zero at $T = 0$, consistent with the expectation from Eq.2 and the existence of the horizontal line node in the gap on α and β [25]. In contrast, we show in Fig. 1 that for the non-chiral (b) pairing state the susceptibility for a-axis B-fields decreases approximately linearly towards $\chi_n/2$ at $T = 0$, again consistent with the expectations of Eq. 4. These results confirm that in spite of very complicated character of the order parameter (see ref. [25]) having zeros on the α and β sheets of the Fermi surface, the overall behavior of the calculated susceptibility tensor is consistent with the expectations from superfluid ^3He [6, 12].

In zero magnetic field, but in the presence of non-zero spin-orbit interaction λ the model predicts that the ground state is the chiral state (a) with d-vector along the crystal c - axis, provided that the spin-orbit coupling leads to a small spin dependence of the effective pairing interaction [11]. Choosing $U' \equiv U^{\uparrow\downarrow}$ about 1% larger than $U \equiv U^{\uparrow\uparrow} = U^{\downarrow\downarrow}$ is sufficient to stabilize the chiral state even for large spin orbit coupling. In contrast, for spin-independent interactions, $U' = U$, the alternative pairing states (b) and (c) ((d) and (e) [11]) are the ground states for any value of $\lambda < 0$ ($\lambda > 0$) [26].

PHASE DIAGRAM IN A MAGNETIC FIELD

In Fig. 2 we show the free energies of the (a) (b) and (c) symmetry pairing states as a function of external magnetic field both for $H \parallel c$ and $H \perp c$. The model parameters were chosen to make the chiral state (a) stable at zero field. For the field in ab-plane the free energy of the chiral state (a) increases with field more slowly than for the (b) and (c) states. This means that in large in-plane fields the (a) state becomes relatively more stable than (b) or (c). On the other hand, in an c-axis field the chiral phase increases its free energy much faster than the other phases, until at a certain critical field, B_t, the (b) or (c) solutions become more stable. Therefore at the field B_t we expect a "spin flop" type phase transition from a d-vector oriented along the c-axis to one where the d-vector lies in the $a - b$ plane. For the parameter values used in Fig. 2 this critical field is $B_t \approx 20$ mT. to one where the d-vector lies in the $a - b$ plane. For the parameter values used in

This prediction is consistent with what one expects from the analogous superfluid state in ^3He-A. In bulk

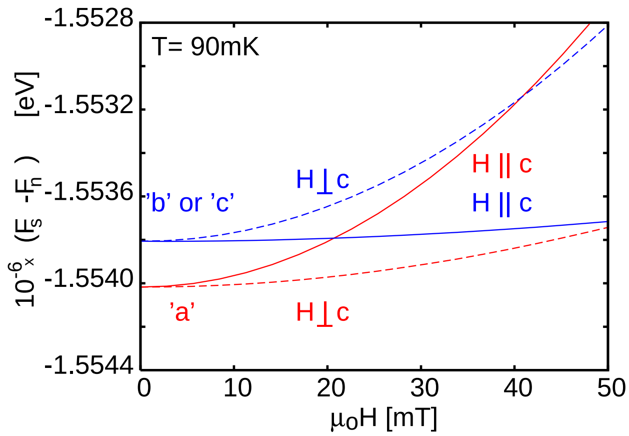

FIG. 2: (Color online) Condensation free energy at $T = 90$mK for three triplet order parameters of different symmetries, (a), (b) and (c) as a function of external field $H \parallel c$ (solid lines) and $H \perp c$ (dashed lines), respectively. The parameters used for calculations are: $\lambda = -0.02t$ where $t = 80$meV is the effective γ band nearest neighbor hopping integral, and $U' = 1.0011U$.

^3He there is no preferred symmetry direction in space and so the d-vector simply rotates continuously to remain perpendicular to the applied field. But in thin films the d-vector is pinned, and only rotates at a finite critical field, known as the Freedericksz transition [27]. In the case of Sr_2RuO_4 the transition is not simply a rotation of the chiral d-vector but is also a transition from a chiral to non-chiral pairing state of different symmetry. The two distinct solutions shown in Fig. 2 (note that (b) and (c) are essentially degenerate) have different entropies and hence this Freedericksz-like spin-flop is a first order thermodynamic phase transition.

One can also ask whether the specific case of Fig 2 is typical for a general set of model parameters. In our effective Hubbard model the spin-orbit interaction enters the Hamiltonian both directly *via* λ in Eq. (5), and also in the spin dependent pairing potential U'/U. These are not really independent, since a full theory, such as a spin fluctuation model [28, 29, 30], would include both effects on the same footing. In Fig. 3 we show a part of the full phase diagram of our model [11] for a specific choice of the parameters λ and U'/U. The inset shows the effect of the c-axis magnetic field in shifting the phase boundary between the chiral state (a) and the alternative (b) and (c) states. At any point in the phase diagram where (a) is stable for zero field, there is a definite critical field for which the spin flop to (b) or (c) takes place. However it is only a small region of the phase diagram, close to the $H = 0$ phase boundary, where the transition takes place in fields as small as $20 - 40$ mT. The implications of the Murakawa *et al.* experiment [7] are that the parameters do indeed lie in this region. The alternative, is that the

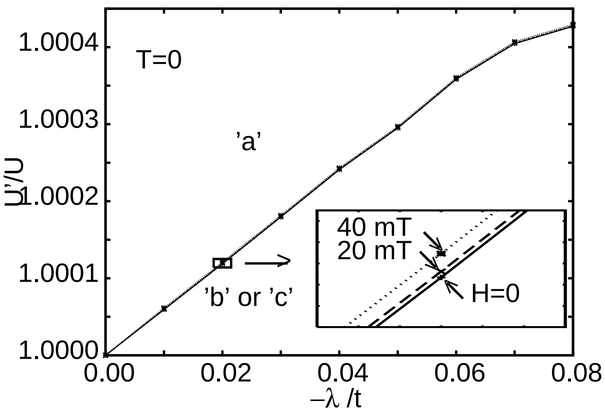

FIG. 3: Critical interaction U'/U as a function of λ in the presence of external field $H \parallel c$ at $T = 0$. The critical c-axis field of the (a) to (b) or (c) spin flop transition depends on how close the physical parameters lie to the zero-field $(a) - (b)/(c)$ phase boundary.

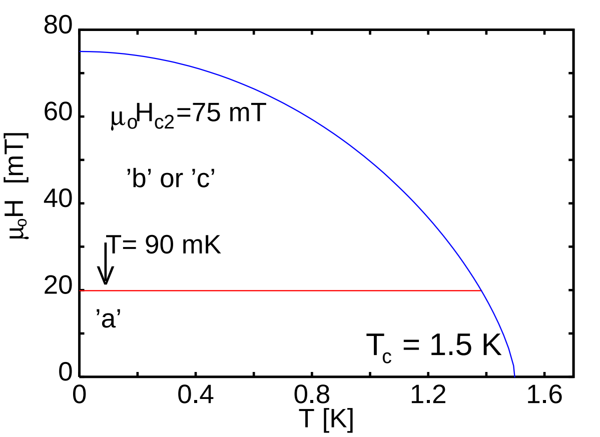

FIG. 4: (Color online) The expected phase diagram in presence of external field $H \parallel c$ in the region below H_{c2}.

spin flop only takes place at a much larger field, and thus would never be observed in any c-axis field below $H_{c2} = 75$ mT, in contradiction to the simplest interpretation of Murakawa *et al.*'s result.

If the relevant parameters do allow the d-vector rotation to occur in a relatively low field, then we expect a phase diagram similar to Fig. 4. This hypothesis makes a definite and clear testable prediction, namely the existence of a new first order phase boundary in the $H - T$ plane, which has not yet been observed. A phase diagram of the type shown in Fig. 4 is required from the experiments [7, 31]. The NQR experiments [7] indicate

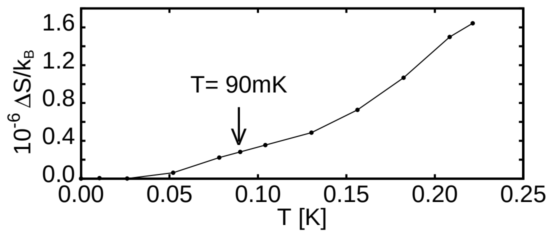

FIG. 5: The temperature dependence of the entropy jump along the transition line $B = B_t = 20$ mT and $\lambda = -0.02t$.

the state with d-vector lying in the (ab) plane is realized in a c-axis magnetic field of order $B = 20$ mT and larger, and that this d-vector orientation must be present at these fields for all temperatures below T_c. On the other hand the nonzero Kerr signal obtained at fields around 10 mT [31] (see the points in Fig. (3a) of this reference and the corresponding discussion in the text) is a clear indication that at these fields the chiral state is stable. Accepting the usual interpretations of these two experiments one is forced to accept the general structure of the phase diagram as in Fig. 4 as the simplest picture which is consistent with both experiments, with a possible margin of uncertainty of about 10 mT in the position of the $B = B_t$ transition line.

An obvious question is why has the predicted phase transition in Fig. 4 not been observed experimentally? Experimental data [32] show multiple superconducting phases at the magnetic field parallel to the (ab) plane, but none have been reported for fields along the c-axis. The answer to this question we suggest is that the phases in question have essentially identical values of the quasiparticle energy gap, $|\mathbf{d}(\mathbf{k})|$ on the Fermi surface, and therefore to a very high level of accuracy they have essentially identical entropy. This is because to leading order only the direction of $\mathbf{d}(\mathbf{k})$ changes at each point on the Fermi surface, and not its magnitude. To confirm this we have calculated the entropy change along the transition line $B = 20$ mT and this is shown in the Fig. 5. The total change in entropy is very small, of order $10^{-6}k_B$ per formula unit, suggesting that it might not have been detected in specific heat experiments. The entropy change in Fig. 5 corresponds to a latent heat of at most 0.003mJ/mol, compared to the zero field specific heat jump $\Delta C/T$ of about 28mJ/K^{2}mol at $T_c = 1.5$K [22]. Assuming that lattice strain or inhomogeneity leads to some smearing of the ideal first order phase transition this small entropy change could easily be masked by the experimental noise.

We should note that in the present calculations we have included the influence of the magnetic field in the Zeeman energies only and neglected the orbital effects altogether. This means that the vortex contribution to the condensation energy of both the (a) and (b) phases is assumed to be the same. There is no reason to expect a significantly

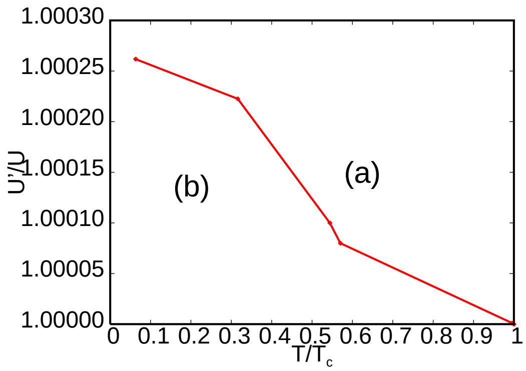

FIG. 6: (Color online) Temperature dependence of the spin dependent interaction anisotropy ratio U'/U necessary for $\lambda = -0.02t$ in the presence of external c-axis field ($H \parallel c$) to stabilize the chiral phase. The symbols (a) and (b) denote the stability regions of corresponding phases.

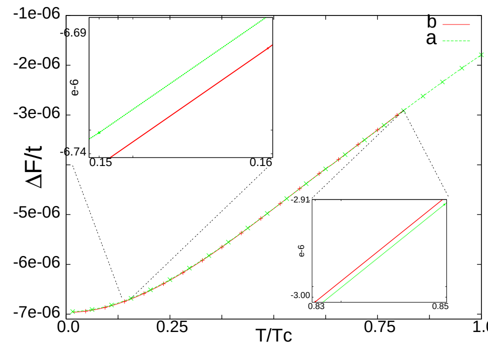

FIG. 7: (Color online) The temperature dependence of the condensation free energies for states 'a' and 'b' in a c-axis magnetic field $B = 30$ mT for $\lambda = -0.02t$ and $U' = 1.0001U$. The insets show the data on the expanded scale for temperature well below (upper left) and above crossing point (lower right).

different vortex lattice response in the different phases, since the only changes are in the direction of $\mathbf{d}(\mathbf{k})$ on the Fermi surface and not in its magnitude $|\mathbf{d}(\mathbf{k})|$.

CONSTRAINTS ON MODEL PARAMETERS

As mentioned above, the experiments [7, 31] indicate that the region where the chiral (a) state is stable is restricted to the low field part of the $H-T$ plane below a critical field which must be in the range of about $B_t \approx 10-20$ mT. This places a number of constraints on the possible parameter values which we can use in our model Hamiltonian.

Using our model Hamiltonian, Eq. 5, we can ask what are the possible values of the spin-dependent pairing interaction needed to qualitatively describe both experiments. In our model this is the ratio $U'/U \equiv U^{\uparrow\downarrow}/U^{\uparrow\uparrow}$ for opposite spin pairing compared to equal spin pairing. The other model parameters are fixed beforehand, including the known band structure and the values of interaction parameters $U_{\parallel}$ and $U_{\perp}$ which were previously fitted to other experiments [20]. For simplicity we have taken the spin-orbit coupling to have a fixed value, $\lambda = -0.02t$, where $t = 80meV$ is the γ band in-plane nearest neighbor hopping integral in our tight binding band structure fit.

With all other parameters fixed, we then calculate the minimum spin dependent interaction enhancement U'/U required to stabilize the chiral state in magnetic fields of up to $B = 20$ mT. It turns out that this minimal value of U'/U changes with temperature, as shown in Fig. 6.

The temperature dependence of U'/U shown in Fig. 6 arises because the condensation energies calculated for a given value of U'/U for (a) and (b) states cross as function of temperature as shown in Fig. 7. For a small value of U'/U for which the (b) state will be stable at low temperatures, we find that this state is destabilized relative to the chiral state closer to T_c.

Let us stress that the model we are using is, in principle at least, valid for arbitrarily large spin-orbit coupling. However, the parametrization of the Fermi surface which we have used ceases to be valid for $|\lambda| > 0.1t$. Therefore in order not to refit the normal state band structure for every value of λ we have limited the calculations to small values of λ. In fact most of the calculations presented in this work have been done for $\lambda = -0.02t$. This restricted choice does not limit the validity of our conclusions, because for every value of the spin-orbit coupling parameter, λ, it is possible to find a corresponding value of U'/U which will stabilize the chiral state at $T = 0K$ as discussed earlier (*c.f.* Fig. 4). Close enough to this boundary of stability a c-axis magnetic field will rotate the d-vector and stabilize one of the other four states [26] independently of the sign or magnitude of λ. This minimal value of U'/U depends not only on λ and B but also on temperature, as shown in Fig. 6. However, this uncertainty in the value of U'/U precludes an unique theoretical determination of the phase diagram on the $H-T$ plane without additional assumptions.

Assuming a constant (*i.e.* temperature independent) value of U'/U which leads to stabilization of the chiral phase at the lowest temperatures would lead to a H-T phase diagram with the chiral phase occupying most of it. The non-chiral state would be limited to low temperature-high field corner. Other assumptions about the temperature dependence of U'/U could produce different topology phase diagrams. Rather than try to explore all these possibilities we simply assume that the boundary between the stable phases at low c-axis fields is given by a line at approximately $B = B_t = 20$ mT (shown in Fig. 2) as dictated by experiment. From this assumption we have then calculated the corresponding temperature dependence of the minimal anisotropy U'/U which is necessary to stabilize the a-phase along the line (as shown in Fig. 6). We find this approach more direct than trying to determine the expected temperature dependence of U'/U *a priori* from a spin fluctuation feed-

back mechanisms [33] stabilizing chiral phase.

We note that our effective pairing Hubbard model does not really include the full self-consistent effects of spin fluctuation feedback [33], which could make the effective pairing interactions U and/or U' temperature dependent. However, the predicted phase transition places severe constraints on the superconducting pairing mechanism when one takes into account a sizable spin-orbit coupling and the detrimental effect of the c-axis B-field on the chiral phase.

SUMMARY AND CONCLUSIONS

To summarize, we have explored the role played by spin-orbit coupling in determining the superconducting states of strontium ruthenate in the presence of c-axis and ab-plane oriented magnetic fields. We showed that the d-vector rotation can provide a consistent interpretation of both the NQR and Kerr effect data, provided the spin-orbit coupling constant λ is small enough. Unfortunately, LDA estimates of λ are significantly larger than the values we have assumed in the calculations presented here[36]. A large value of spin-orbit coupling has previously been cited as evidence against the d-vector rotation picture [34, 35, 36]. Despite these objections, it is clear from our results that even for large values of λ the transition field B_t for d-vector rotation, in Fig. 2, can lie within experimental constraints [7, 31], but only if the pairing interaction spin anisotropy U'/U is such as to make the free energy difference between (a) and (b) phases finely balanced so that a small field of order 20 mT is sufficient to cause the d-vector rotation. We have shown that for any value of λ there is corresponding anisotropy U'/U such that this balance can be achieved. It is possible that such balancing is a consequence of a spin-fluctuation feedback mechanism [33], or emerges directly from the full microscopic spin fluctuation theory. But in the absence of a detailed theory of this effect further progress in the field must await experimental measurement of the effective spin-orbit parameter for the quasiparticles at the Fermi surface.

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